TITLE OF THE INVENTION

PROCESS FOR THE SURFACE-IMMOBILIZATION OF ANTI-MICROBIAL POLYMERS BY METAL DEPOSITION

CROSS-REFERENCE TO RELATED APPLICATIONS

The present application claims priority to German Application No. DE 102 384 85.1, filed on August 22, 2002, which is hereby incorporated by reference in its entirety.

BACKGROUND OF THE INVENTION

Field of the Invention

The present invention relates to a process for the surface-immobilization of antimicrobial polymers, where the surface-immobilization of the antimicrobial polymers takes place by metal deposition, and also to a metal coating having antimicrobial properties.

Discussion of the Background

The surfaces of pipelines, containers, and packaging are susceptible to undesirable colonization and propagation of bacteria. Coats of slime can form on these surfaces, which give rise to extremely high levels of microbial populations. This phenomenon can adversely affect the quality of water, beverages, and foods intended for human consumption because it causes these products to decay. Therefore, it may even damage the health of consumers.

Good hygiene is important for products intended for human consumption or intimate human contact, including the treatment, prevention, and reduction of bacterial growth on these products. These products may include textiles, especially those textiles intended for use near and around the genital area of individuals. Further, good hygiene is required for textiles required in the care of the sick and the elderly.

Good hygiene is required in and around hospitals. This includes hospital wards, areas for medical interventions, and toilets. Examples of hospital wards include but are not limited to intensive care, neonatal and isolation wards. Isolation wards include those in which critical cases of infection are treated. There is a need for bacteria to be kept away from all surfaces, such as surfaces of furniture and instruments, in and around hospitals.

The growth of microbes may also adversely affect many industrial systems. In particular, separating materials, which utilize membranes or filters are severely impaired by the deposition and growth of microbes. In seawater desalination the growth of marine algae in the system may shorten running times, while the growth of biofilms may prematurely block the filter cake in deep-bed filtration. To counter the growth of biofilms, crossflow filtration has been employed. Crossflow filtration utilizes a specified flow perpendicular to the plane of filtration. However, this method has proven to be industrially inadequate.

At present, surfaces of furniture, textiles, and equipment are commonly treated with chemicals or solutions with broad and general antimicrobial activity to prevent bacterial colonization. These general antimicrobial chemical agents act nonspecifically and frequently act as a human irritant or are either directly toxic or its degradation products are toxic. An additional problem associated with these broadly nonspecific antimicrobial chemicals is an increased intolerance among humans arising from frequent contact. Another procedure to counteract the spread of bacteria on surfaces is by incorporating antimicrobial substances into a matrix.

Another challenge of constantly increasing significance is the prevention of algal growth on surfaces. This challenge is underscored by the quantity of external surfaces on buildings with plastic cladding, which is particularly susceptible to colonization by algae. In addition to an undesirable appearance, in some circumstances algae growth on surfaces can also impair the functions of the components concerned. A relevant example of this effect is surface colonization by algae which has a photovoltaic function.

Another form of microbial contamination, for which again no technically satisfactory solution has been found, is fungal infestation of surfaces. For example, *Aspergillus niger* infestation of joints or walls in wet areas not only impairs appearance, but also has serious health implications. Owing to the frequency of allergic response substances released by the fungi, serious, chronic respiratory diseases may result.

In the marine sector, fouling of the hulls of boats affects costs, since the growth of the fouling organism is correlated with an increase in the boat's flow resistance, and thus a marked increase in fuel consumption. Problems of this type have hitherto generally been countered by incorporating toxic heavy metals or other low-molecular-weight biocides into antifouling coatings in an attempt to mitigate the costs associated with increased fuel consumption. To this end, the damaging side effects of coatings of this type are generally accepted, but as society's environmental awareness rises this state of affairs becomes increasingly problematic.

US Patent 4,532,269 describes a terpolymer made from butyl methacrylate, tributyltin methacrylate, and tert-butylaminoethyl methacrylate. This copolymer is used as an antimicrobial paint for ships; however, the hydrophilic tert-butylaminoethyl methacrylate promotes slow erosion of the polymer and a corresponding release of a highly toxic tributyltin methacrylate as active antimicrobial agent. In these applications, the copolymer prepared using amino methacrylates is merely a matrix or carrier for added microbicidal active ingredients which may diffuse or migrate out of the carrier material. At some stage polymers of this type lose their activity once the necessary minimum inhibitor concentration (MIC) at the surface has been lost.

European Patent Application 0 862 858 describes copolymers of tert-butylaminoethyl methacrylate, and a methacrylate having a secondary amino function, which have inherent microbicidal properties. However, a particular problem which arises in the industrial application of antimicrobial polymers is an ability to permanently immobilize these polymers on surfaces. As a result of erosion, abrasion, and swelling behavior of these polymers, durable adhesion to surfaces has so far proven to be an unsolved problem. The German Patent Application DE 101 49 973 (*unpublished at the date of this patent application*) describes a process for preparing extraction-resistant coatings of antimicrobial polymers by minimizing the coating thickness, but even this process cannot completely suppress swelling and the associated impairment through partial separation of surface constituents.

Accordingly, it is highly desirable to find a process by which an antimicrobial polymer may be immobilized on surfaces where the surface-immobilized antimicrobial polymer does not leach off from the surface and is free of the other disadvantages described above.

SUMMARY OF THE INVENTION

It is an object of the present invention to provide a process by which an antimicrobial polymer may be immobilized on surfaces where the surface-immobilized antimicrobial polymer does not leach off from the surface and is free of the other disadvantages described above.

In a preferred object is a process for the surface-immobilization of antimicrobial polymers, comprising:

forming a process bath comprising at least one antimicrobial polymer; and

surface-immobilize the antimicrobial polymers to a surface of a workpiece by coating the workpiece by metal deposition.

Within this object the metal deposition may electrochemical metal deposition and may be with or without an external current. This process may entail immersing the workpiece in the process bath for a time and under conditions suitable for forming a metal layer of a desired thickness.

In another object is a metal coating which comprises one or more antimicrobial polymers, wherein the surface of the metal coating comprises from 0.1 to 20% by surface area of said antimicrobial polymers.

In a further object of the present invention the metal coating is produced by a process, comprising:

forming a process bath comprising at least one antimicrobial polymer; and

surface-immobilize the antimicrobial polymers to a surface of a workpiece by coating the workpiece by metal deposition.

In yet another object is a building, a monument, or a galvanic cell containing a coated workpiece produced in accordance with the previously specified inventive method.

The above objects highlight certain aspects of the invention. Additional objects, aspects and embodiments of the invention are found in the following detailed description of the invention.

DETAILED DESCRIPTION OF THE INVENTION

Unless specifically defined, all technical and scientific terms used herein have the same meaning as commonly understood by a skilled artisan in biochemistry, cellular biology, molecular biology, and the medical sciences.

All methods and materials similar or equivalent to those described herein can be used in the practice or testing of the present invention, with suitable methods and materials being described herein. All publications, patent applications, patents, and other references mentioned herein are incorporated by reference in their entirety. In case of conflict, the present specification, including definitions, will control. Further, the materials, methods, and examples are illustrative only and are not intended to be limiting, unless otherwise specified.

The present invention is based, in part, on the inventor's discovery that permanent and stable surface-immobilization of antimicrobial polymers can be obtained by metal deposition and complies with the requirements profile described in an almost ideal fashion. Metal

coatings deposited electrochemically, with or without the application of an external current, have proven successful in improving the wear performance of the surfaces of materials subjected to mechanical stress, and these coatings now have a wide variety of industrial uses. If the antimicrobial polymers, which are present homogeneously distributed in the process bath, are uniformly embedded into the metal coating the result is durable surface-adhesion of the antimicrobial polymers to the, or within the, metal coating.

The present invention therefore provides a process for the surface-immobilization of antimicrobial polymers, where the surface-immobilization of the antimicrobial polymers takes place by metal deposition. The invention further provides a metal coating made thereby, which has anti-microbial properties.

The process for the surface-immobilization of anti-microbial polymers comprises using metal deposition, which takes place either without external current or by means of external current, for the surface-immobilization of the antimicrobial polymers.

In the process of the invention, the metal deposition preferably takes place from a process bath having at least one antimicrobial polymer alongside constituents known to the skilled artisan. The workpiece to be coated can be immersed partially or entirely into the process bath for a time and under conditions suitable to achieve the desired thickness of the metal coating layer. To obtain uniform embedding of the antimicrobial polymers in the metal coating, the antimicrobial polymers preferably have a very fine prior distribution and dispersion in the process bath.

In the process of the invention, the antimicrobial polymer is preferably therefore added in the form of an aqueous dispersion. This aqueous dispersion preferably contains from 0.01 to 30% by volume, preferably from 0.1 to 10% by volume, particularly preferably from 0.3 to 1% by volume, of the antimicrobial polymer. As an alternative to the use of a dispersion of the antimicrobial polymer, it is also in principle possible for these to be introduced in the form of very fine suspended polymer particles. According to this invention, furthermore, nitrogen-containing antimicrobial polymers may often be converted into more soluble forms, which are therefore more suitable for the process, by addition of acid and the associated partial or quantitative protonation of the nitrogen atoms.

Depending on the amount of antimicrobial polymers added to the process bath during the process of the present invention, and on the particle size of these polymers, and also on the selected hydrodynamic conditions during the deposition procedure, it is possible to achieve various maximum embedding volumes of the antimicrobial polymer in the metal coatings.

The antimicrobial polymers are preferably prepared from at least one nitrogen-functionalized monomers or phosphorus-functionalized monomers. Antimicrobial polymers particularly suited for this purpose are those prepared from at least one monomer from the group consisting of 2-tert-butylaminoethyl methacrylate, 2-diethylaminoethyl methacrylate, 2-diethylaminomethyl methacrylate, 2-tert-butylaminoethyl acrylate, 3-dimethylaminopropyl acrylate, 2-diethylaminoethyl acrylate, 2-dimethylaminoethyl acrylate, dimethylamino-propylmethacrylamide, diethylaminopropylmethacrylamide, N-3-dimethyl-aminopropylacrylamide, 2-methacryloyloxy-ethyltrimethylammonium methosulfate, 2-methacryloyl-oxyethyltrimethylammonium chloride, 3-methacryloyl-aminopropyl-trimethylammonium chloride, 2-acryloyloxy-ethyl-4-benzoylbenzyldimethylammonium bromide, 2-methacryloyloxyethyl-4-benzoylbenzyldimethylammonium bromide, allyltriphenylphosphonium bromide, allyltriphenylphosphonium chloride, 2-acrylamido-2-methyl-1-propanesulfonic acid, 2-diethylaminoethyl vinyl ether, 3-aminopropyl vinyl ether, 3-aminopropyl methacrylate, 2-aminoethyl methacrylate, 4-aminobutyl methacrylate, 5-aminopentyl methacrylate, 3-aminopropyl acrylate, 2-aminopropyl acrylate, 4-aminobutyl acrylate, 5-aminopentyl acrylate, 2-aminoethyl vinyl ether, 4-aminobutyl vinyl ether, and 5-aminopentyl vinyl ether.

In one particular embodiment of the present invention, the antimicrobial polymers may be prepared using an additional aliphatically unsaturated monomers in addition to the monomers mentioned. These other aliphatically unsaturated monomers do not necessarily have to have any additional antimicrobial action. Monomers suitable for this purpose are acrylic or methacrylic compounds, e.g. acrylic acid, tert-butyl methacrylate, methyl methacrylate, styrene or its derivatives, vinyl chloride, vinyl ethers, acrylamides, acrylonitriles, olefins (ethylene, propylene, butylene, isobutylene), allyl compounds, vinyl ketones, vinylacetic acid, vinyl acetate, or vinyl esters, ethyl methacrylate, butyl methacrylate, methyl acrylate, ethyl acrylate, butyl acrylate, and tert-butyl acrylate.

The antimicrobial polymers used may have molar masses ranging from 5 000 to 5 000 000 g/mol, in particular from 20 000 to 1 000 000 g/mol, preferably from 50 000 to 500 000 g/mol (weight average).

In another embodiment of the present invention, antimicrobial polymers may be prepared using a polymer blend made from antimicrobial and non-antimicrobial polymers. Examples of the non-antimicrobial polymers include polymethyl methacrylate, PVC, polyacrylic acid, polystyrene, polyolefins, polyterephthalates, polyamides, polysulfones, polyacrylonitrile, polycarbonates, polyurethane, and cellulose derivatives.

The metals deposited by the metal deposition step of the inventive process are preferably nickel, copper, silver, gold, platinum, or alloys of these metals, particularly preferably nickel or copper.

In the process of the invention, the metal deposition may take place without external current and be based on reductive precipitation of metal on the material to be coated. The skilled artisan may reduce the metal ions by two primary means:

- deposition through charge exchange in a coating process using a less noble metal (immersion or dipping),

- deposition by a reductive process where the electrons needed to reduce the metal ions are produced with the aid of a chemical reducing agent whose standard potential has to be substantially more negative than that of the metal to be deposited. The reducing agent is oxidized and the electrons released reduce the metal ions and result in metal deposition.

A third alternative exists for reducing metal ions, deposition by bringing the primary metal into contact with a third metal which functions as electron donor, is little used in industry.

The antimicrobial polymer, surface-immobilization process of the present invention by metal deposition without external current preferably is conducted by a reductive process. For this process, the process bath preferably contains from 0.01 to 30% by volume, particularly preferably from 5 to 15% by volume, very particularly preferably from 8 to 12% by volume, of an aqueous dispersion of antimicrobial polymer. In addition to the antimicrobial polymer this process bath comprises, for the particular embodiment of metal deposition without external current, at least one metal salt of the metal to be deposited, with preference being given to acetate, halides, or sulfates of the metal to be deposited.

This process bath for metal deposition without external current may also contain a reducing agent, preferably sodium hypophosphite, sodium borohydride, alkali metal amino-boranes, or formaldehyde. The process bath may also contain a complexer, preferably oxycarboxylic acids, particularly preferably citric acid, glycine, ethylenediamine, ethylenediaminetetraacetic acid (EDTA), potassium sodium tartrate, or ethylenediamine propoxylate.

The metal deposition of the process of the present invention, without external current, preferably is conducted at a pH of from 2 to 12, in particular at a pH of from 2 to 7. To ensure constant pH in the process bath, so that the deposition process is not impaired by pH change, it is preferable to add buffer substances to the process bath, in particular organic acids and alkali metal salts of these.

An advantage provided by metal deposition of the process of the invention, without external current, is uniform metal deposition with simultaneous binding of the antimicrobial polymer to the surface of the entire workpiece, including, for example, in the interior of a tube, since the reduction of the metal ion proceeds independently of the shape of the workpiece or of the anode. The same thickness of metal layer is thus obtained at all sites on the workpiece.

In another embodiment of the process of the invention, the metal deposition may take place by means of external current, preferably in an electrolysis cell, particularly preferably in a glass cell. It can be advantageous for the metal deposition to be carried out in an electrolysis cell or glass cell which can be thermostatically controlled.

In the case of metal deposition of the process of the invention by means of external current, the workpiece to be coated is preferably a cathode, the anode preferably being graphite or the metal which is to be deposited on the workpiece to be coated.

The process bath, in this case the electrolysis bath, of the process of the invention preferably comprises from 0.01 to 30% by volume, particularly preferably from 5 to 15% by volume, very particularly preferably from 8 to 12% by volume, of the aqueous dispersion of the antimicrobial polymer, alongside the metal salt, which is preferably the sulfate or halide of the metal to be deposited on the workpiece.

If the metal of the anode material is not soluble in the electrolysis bath, it can be advantageous to add more metal salt to the electrolysis bath at regular intervals.

This invention also provides a metal coating which has antimicrobial properties. The surface of this metal coating of the invention comprises from 0.1 to 20% by surface area, preferably from 0.2 to 15% by surface area, particularly preferably from 0.5 to 10% by surface area, of antimicrobial polymers.

The antimicrobial polymers of the metal coating of the invention are preferably prepared from nitrogen-functionalized monomers or phosphorus-functionalized monomers. Antimicrobial polymers particularly suited for this purpose are those prepared from at least one monomer from the group consisting of 2-tert-butylaminoethyl methacrylate, 2-diethyl-aminoethyl methacrylate, 2-diethylaminomethyl methacrylate, 2-tert-butylaminoethyl

acrylate, 3-di-methylaminopropyl acrylate, 2-diethylaminoethyl acrylate, 2-dimethyl-aminoethyl acrylate, dimethylamino-propylmethacrylamide, diethylaminopropylmeth-acrylamide, N-3-dimethylaminopropylacrylamide, 2-methacryloyloxy-ethyltrimethyl-ammonium methosulfate, 2-methacryloyl-oxyethyltrimethylammonium chloride, 3-methacryloyl-aminopropyltrimethylammonium chloride, 2-acryloyloxy-ethyl-4-benzoyl-benzyldimethylammonium bromide, 2-methacryloyloxyethyl-4-benzoylbenzyldimethylammonium bromide, allyltriphenylphosphonium bromide, allyltriphenylphosphonium chloride, 2-acrylamido-2-methyl-1-propanesulfonic acid, 2-diethylaminoethyl vinyl ether, 3-aminopropyl vinyl ether, 3-aminopropyl methacrylate, 2-aminoethyl methacrylate, 4-aminobutyl methacrylate, 5-aminopentyl methacrylate, 3-aminopropyl acrylate, 2-aminopropyl acrylate, 4-aminobutyl acrylate, 5-aminopentyl acrylate, 2-aminoethyl vinyl ether, 4-aminobutyl vinyl ether, and 5-aminopentyl vinyl ether.

In one particular embodiment of the present invention, the antimicrobial polymers may be prepared using an additional aliphatically unsaturated monomers in addition to the monomers mentioned. These other aliphatically unsaturated monomers do not necessarily have to have any additional antimicrobial action. Monomers suitable for this purpose are acrylic or methacrylic compounds, e.g. acrylic acid, tert-butyl methacrylate, methyl methacrylate, styrene or its derivatives, vinyl chloride, vinyl ethers, acrylamides, acrylonitriles, olefins (ethylene, propylene, butylene, isobutylene), allyl compounds, vinyl ketones, vinylacetic acid, vinyl acetate or vinyl esters, ethyl methacrylate, butyl methacrylate, methyl acrylate, ethyl acrylate, butyl acrylate, and tert-butyl acrylate.

The antimicrobial polymers used may have molar masses of from 5 000 to 5 000 000 g/mol, in particular from 20 000 to 1 000 000 g/mol, preferably from 50 000 to 500 000 g/mol (weight average).

In another embodiment of the metal coating of the invention, this comprises antimicrobial polymers prepared using a polymer blend made from antimicrobial and non-antimicrobial polymers. Examples of these non-antimicrobial polymers are polymethyl methacrylate, PVC, polyacrylic acid, polystyrene, polyolefins, polyterephthalates, polyamides, polysulfones, polyacrylonitrile, polycarbonates, polyurethane, and cellulose derivatives.

The metal coating of the present invention are those that are preferably produced by the process of the present invention.

Once the workpieces are treated according to the present invention, they may be used in the protection of the surfaces used in construction of buildings, monuments, or galvanic cells.

Having generally described this invention, a further understanding can be obtained by reference to certain specific examples, which are provided herein for purposes of illustration only, and are not intended to be limiting unless otherwise specified.

EXAMPLES

Example 1:

16 mL of tert-butylaminoethyl methacrylate (Aldrich), 45 g of Triton X 405 (Aldrich), 200 mL of deionized water and 0.6 g of potassium peroxodisulfate (Aldrich) were charged to a three-necked flask and heated to 60°C under a stream of argon. A further 180 mL of tert-butylaminoethyl methacrylate were then added dropwise over a period of 4 hours. The mixture was then stirred at 60°C for a further 2 hours, and the resultant emulsion was then allowed to cool to room temperature.

Example 1a:

The layer was formed in a glass cell which was thermostatically controlled and had a volume of 200 mL at a temperature of 55°C, on a rod electrode made from titanium. The anode used comprises a rotationally symmetrical nickel cylinder. The electrolyte solution was composed of 150 mL of a solution of 220 g/L of $NiSO_4*7H_2O$, 18 g/L of $NiCl_2*6H_2O$, 18 g/L of H_3BO_3, and also 15 mL of the product from example 1. The pH was then adjusted to 3.5 by adding sulfuric acid and the cathodic current density was adjusted to about 10 A/dm^2. The experiment was terminated after 30 minutes and the titanium electrode was removed.

Example 1b:

The layer was formed in a glass cell which was be thermostatically controlled and had a volume of 200 mL at a temperature of 55°C, on a rod electrode made from stainless steel. The anode used comprises a rotationally symmetrical nickel cylinder. The electrolyte solution was composed of 150 mL of a solution of 220 g/L of $NiSO_4*7H_2O$, 18 g/L of $NiCl_2*6H_2O$, 18 g/L of H_3BO_3, and also 15 mL of the product from example 1. The pH was then adjusted to 3.5 by adding sulfuric acid and the cathodic current density was adjusted to about 10 A/dm^2. The experiment was terminated after 30 minutes and the stainless steel electrode was removed.

Example 1c:

The coated electrode from example 1a was held on the base of a glass beaker in which there were 10 mL of a test microbial suspension of Pseudomonas aeruginosa. The resultant system was then shaken for 4 hours. 1 mL of the test microbial suspension was then removed. After expiry of this time, the number of microbes had fallen from 10^7 to 10^4 microbes per mL.

Example 1d:

The coated electrode from example 1b was held on the base of a glass beaker in which there were 10 mL of a test microbial suspension of Pseudomonas aeruginosa. The resultant system was then shaken for 4 hours. 1 mL of the test microbial suspension was then removed. After expiry of this time, the number of microbes had fallen from 10^7 to 10^4 microbes per mL.

Example 1e:

The coated electrode from example 1a was held on the base of a glass beaker in which there were 10 mL of a test microbial suspension of Staphylococcus aureus. The resultant system was then shaken for 4 hours. 1 mL of the test microbial suspension was then removed. After expiry of this time, the number of microbes had fallen from 10^7 to 10^3 microbes per mL.

Example 1f:

The coated electrode from example 1b was held on the base of a glass beaker in which there were 10 mL of a test microbial suspension of Staphylococcus aureus. The resultant system was then shaken for 4 hours. 1 mL of the test microbial suspension was then removed. After expiry of this time, the number of microbes had fallen from 10^7 to 10^3 microbes per mL.

Example 1g:

The surface of the coated electrode from example 1a was roughened with a fine-grain abrasive paper, then placed in water at 60°C for 15 minutes. The electrode thus treated was then held on the base of a glass beaker in which there were 10 mL of a test microbial suspension of Staphylococcus aureus. The resultant system was then shaken for 4 hours. 1 mL of the test microbial suspension was then removed. After expiry of this time the number of microbes had fallen from 10^7 to 10^3 microbes per mL.

Example 1h:

The surface of the coated electrode from example 1b was roughened with a fine-grain abrasive paper, then placed in water at 60°C for 15 minutes. The electrode thus treated was then held on the base of a glass beaker in which there are 10 mL of a test microbial suspension of Staphylococcus aureus. The resultant system was then shaken for 4 hours. 1 mL of the test microbial suspension was then removed. After expiry of this time the number of microbes had fallen from 10^7 to 10^3 microbes per mL.

Example 2:

18 mL of dimethylaminopropylmethacrylamide (Aldrich), 43 g of Triton X 405 (Aldrich), 200 mL of deionized water and 0.5 g of potassium peroxodisulfate (Aldrich) are charged to a three-necked flask and heated to 60°C under a stream of argon. A further 180 mL of dimethylaminopropylmethacrylamide are then added dropwise over a period of 4 hours. The mixture is then stirred at 60°C for a further 2 hours, and the resultant emulsion is then allowed to cool to room temperature.

Example 2a:

The layer is formed in a glass cell which can be thermostatically controlled and has a volume of 200 mL at a temperature of 55°C, on a rod electrode made from titanium. The anode used comprises a rotationally symmetrical nickel cylinder. The electrolyte solution is composed of 150 mL of a solution of 220 g/L of $NiSO_4*7H_2O$, 18 g/L of $NiCl_2*6H_2O$, 18 g/L of H_3BO_3, and also 15 mL of the product from example 2. The pH is then adjusted to 3.5 by adding sulfuric acid and the cathodic current density is adjusted to about 10 A/dm^2. The experiment is terminated after 30 minutes and the titanium electrode is removed.

Example 2b:

The layer is formed in a glass cell which can be thermostatically controlled and has a volume of 200 mL at a temperature of 55°C, on a rod electrode made from stainless steel. The anode used comprises a rotationally symmetrical nickel cylinder. The electrolyte solution is composed of 150 mL of a solution of 220 g/L of $NiSO_4*7H_2O$, 18 g/L of $NiCl_2*6H_2O$, 18 g/L of H_3BO_3, and also 15 mL of the product from example 2. The pH is then adjusted to 3.5 by adding sulfuric acid and the cathodic current density is adjusted to about 10 A/dm^2. The experiment is terminated after 30 minutes and the stainless steel electrode is removed.

Example 2c:

The coated electrode from example 2a is held on the base of a glass beaker in which there are 10 mL of a test microbial suspension of Pseudomonas aeruginosa. The resultant system is then shaken for 4 hours. 1 mL of the test microbial suspension is then removed. After expiry of this time, the number of microbes is expected to have fallen from 10^7 to 10^5 microbes per mL.

Example 2d:

The coated electrode from example 2b is held on the base of a glass beaker in which there are 10 mL of a test microbial suspension of Pseudomonas aeruginosa. The resultant system is then shaken for 4 hours. 1 mL of the test microbial suspension is then removed. After expiry of this time, the number of microbes is expected to have fallen from 10^7 to 10^5 microbes per mL.

Example 2e:

The coated electrode from example 2a is held on the base of a glass beaker in which there are 10 mL of a test microbial suspension of Staphylococcus aureus. The resultant system is then shaken for 4 hours. 1 mL of the test microbial suspension is then removed. After expiry of this time, the number of microbes is expected to have fallen from 10^7 to 10^4 microbes per mL.

Example 2f:

The coated electrode from example 2b is held on the base of a glass beaker in which there are 10 mL of a test microbial suspension of Staphylococcus aureus. The resultant system is then shaken for 4 hours. 1 mL of the test microbial suspension is then removed. After expiry of this time, the number of microbes is expected to have fallen from 10^7 to 10^4 microbes per mL.

Example 2g:

The surface of the coated electrode from example 2a is roughened with a fine-grain abrasive paper, then placed in water at 60°C for 15 minutes. The electrode thus treated is then held on the base of a glass beaker in which there are 10 mL of a test microbial suspension of Staphylococcus aureus. The resultant system is then shaken for 4 hours. 1 mL of the test

microbial suspension is then removed. After expiry of this time the number of microbes is expected to have fallen from 10^7 to 10^4 microbes per mL.

Example 2h:

The surface of the coated electrode from example 2b is roughened with a fine-grain abrasive paper, then placed in water at 60°C for 15 minutes. The electrode thus treated is then held on the base of a glass beaker in which there are 10 mL of a test microbial suspension of Staphylococcus aureus. The resultant system is then shaken for 4 hours. 1 mL of the test microbial suspension is then removed. After expiry of this time the number of microbes is expected to have fallen from 10^7 to 10^4 microbes per mL.

Numerous modifications and variations on the present invention are possible in light of the above teachings. It is, therefore, to be understood that within the scope of the accompanying claims, the invention may be practiced otherwise than as specifically described herein.